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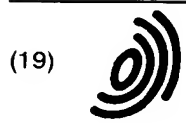
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(54) **Biosoluble composition of glass fibres for the production of glass wools and the like**

(57) The composition includes the following compounds in the specified weight percentages of the whole composition: %SiO₂ 60-70; %Al₂O₃ 1-3; %Na₂O+%K₂O > 17; %CaO ≤ 8 %MgO 2-4%; CaO+%MgO < 10.5; %R₂O/%RO > 1.65; %B₂O₃ 4.5-8; where RO is the sum of oxides of elements pertaining to column 2A of the Periodic Table and R₂O is the sum of oxides of elements pertaining to column 1A of the Peri-

odic Table, the composition further including up to about 2 % in weight of impurities and other conventional compounds such as Fe₂O₃, but P₂O₅ and SO₃ being excluded from the composition. The invention provides biosoluble fibres for glass wools with relatively low KI values.

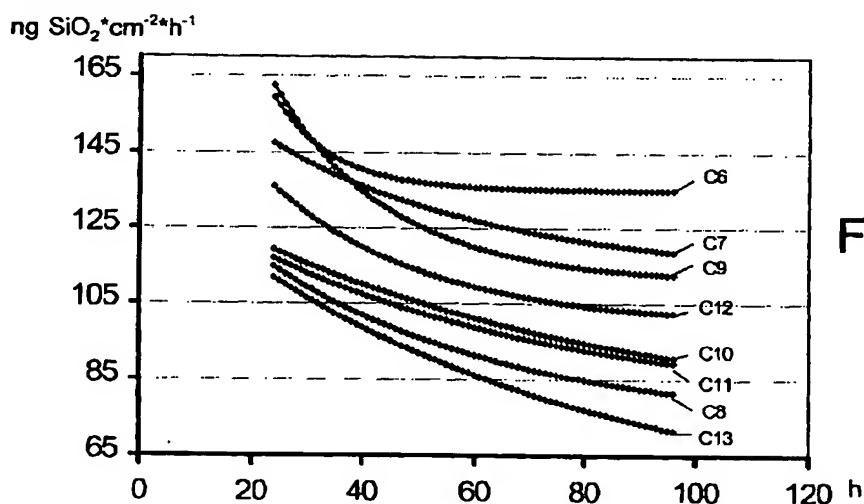


Fig 2

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Description

Technical field to which the invention applies

- 5 [0001] The present invention refers to biosoluble (or biodegradable) compositions of glass fibres for the production of glass wools and the like.

Prior art

- 10 [0002] In the last years, it has been raised that the particles released during the manufacture, storage and use of glass wool products and inhaled by individuals in their environment are potentially cancerogenic if gathered in the lung. Because of this, several standards and recommendations have been introduced to ensure that the fibres with which said glass wools are produced are bioremovable either by macrophage digestion and mucolytic expulsion or by dissolution in the lung body fluids.
- 15 [0003] In order to measure biosolubility of the particles capable of being inhaled, *in vivo* and *in vitro* tests are used. [0004] *In vivo* biosolubility tests attempt to determine the average residence time of particles in the lung by means of different protocols, among which the "*Biopersistence of fibres. Short-term exposure by inhalation*", emanating from the European Community (EC/TM/26 rev. 7, 1998). Its object is to calculate the half-life of the particles in the lung, by obtaining a numerical value showing the above mentioned half-life ($T_{1/2}$) in days.
- 20 [0005] *In vitro* tests are carried out by simulating the action of lung fluid over fibre particles to establish again a numerical value correlating to its presumed half-life ($T_{1/2}$) in the lung. [0006] It is assumed that a fibre is acceptably biosoluble if its half-life is ≤ 10 days in an *in vivo* inhalation test. [0007] Apart from fibre biosolubility tests *in vivo* and *in vitro*, and presumably upon considering that there does not exist an evident and irrefutable correlation between cancer and the gathering of said particles in the lung, the Authorities in Germany introduced (on 12 June 1998) a decree in which the so-called "KI index" ("*Dritte Verordnung zur Änderung der Gefahrstoffverordnung*") is established. This index is defined by the following formula:
- 25

$$KI = \%Na_2O + \%K_2O + \%CaO + \%MgO + \%B_2O_3 + \%BaO - 2 \cdot \%Al_2O_3.$$

- 30 [0008] With $KI \geq 40$, a fibre is considered harmless, without the need to perform confirmatory tests. With $KI < 40$, it may be necessary to confirm the harmless nature of the fibre through biosolubility tests. [0009] The prior art documents which are considered to be most representative to the present invention are the following: EP 0412878 --proposing that the fibre includes more than 0.1 % in weight of phosphorus pentoxide (P_2O_5) when the percentage in weight of alumina (Al_2O_3) is equal to or higher than 1 %--, EP 0738692 and EP 0738693 --proposing high contents of boron oxide (B_2O_3) with the possible use of P_2O_5 --, WO 9634836 --proposing fibres with $KI \geq 40$ --, FR 2758322 --proposing fibres with $KI > 30$, preferably $KI > 35$, and low ratios R_2O/RO (ratio of alkaline oxides/alkaline-earth oxides)--, EP 0872458 and WO 9906332 --proposing relatively low contents of silica (SiO_2) and high KI.
- 35 [0010] The general trend reflected by the majority of the most recent prior art documents is that manufacturers have tried to increase the dissolution capacity of the fibre by decreasing the conventional amount of SiO_2 and Al_2O_3 and by the addition of new oxides, particularly P_2O_5 and sulphur trioxide (SO_3), also attempting to reach KI values ≥ 40 . [0011] However, the approach of the above mentioned trend has obvious disadvantages:
- 40
- the decrease of SiO_2 brings about a decrease in the melt viscosity;
 - 45 - the decrease of Al_2O_3 implies a corresponding decrease in the mechanical strength of the fibres;
 - the presence of P_2O_5 has a negative effect on the fibre strength to mechanical stresses, increases the production of dust and causes that the refractory of the furnaces (especially gas-fired furnaces) for manufacturing the fibre suffers the aggressive attack of P_2O_5 ;
 - the presence of SO_3 determines a greater corrosion of the discs which produce the fibres.
- 50 [0012] With respect to the negative effect of P_2O_5 to the mechanical strength of the fibres, the inventors found through testing that an increase from 0 to 0.8 % of P_2O_5 in detriment, for example, of the same amount of Al_2O_3 , results in a decrease nearly linear to the mechanical strength to 60 % of the original one.

55 Disclosure of the invention

- [0013] The present inventors assigned themselves the task of obtaining new biosoluble wool fibres. Unlike the current manufacturer's trend mentioned above, the experiments of the present inventors were carried out under the follow-

ing general guidelines:

- firstly, the use of P_2O_5 and SO_3 was omitted;
- secondly, relatively high contents of SiO_2 and Al_2O_3 were maintained in order to enhance the viscosity of the melt and the mechanical strength of the fibres;
- thirdly, the amount of RO (sum of oxides of elements pertaining to column 2A of the Periodic Table, and in practice sum %CaO+%MgO) was decreased to improve the workability of the melt, and to avoid its devitrification during the fibre formation; and
- fourthly, the amount of R_2O (sum of oxides of elements pertaining to column 1A of the Periodic Table, and in practice sum %Na₂O+%K₂O) was increased so as to compensate, from the biosolubility viewpoint, for the relatively high content of Al_2O_3 .

[0014] Surprisingly and unexpectedly, the inventors were able to confirm through relevant testing, the biosolubility of the fibres of the invention, in spite of the fact that these fibres have a KI value well below 40 and even below 30. They also confirmed that the fibres of the invention imply a good compromise between, on the one hand, their biosolubility and, on the other hand, their mechanical properties and workability.

[0015] Thereby, the invention refers to a biosoluble composition of glass fibres for the production of glass wools and the like, characterized in that it includes the following compounds in the specified weight percentages of the whole composition:

%SiO ₂	60-70
%Al ₂ O ₃	1-3
%Na ₂ O+%K ₂ O	> 17
%CaO	≤ 8
%MgO	2-4
%CaO+%MgO	< 10.5
%R ₂ O/%RO	> 1.65
%B ₂ O ₃	4.5-8,

where RO is the sum of oxides of elements pertaining to column 2A of the Periodic Table and R_2O is the sum of oxides of elements pertaining to column 1A of the Periodic Table, the composition further including up to about 2 % in weight of impurities and other conventional compounds such as Fe_2O_3 , but P_2O_5 and SO_3 being excluded from the composition.

[0016] According to an optional feature of the invention, the composition includes 60-68 % of SiO_2 and in that %Na₂O+%K₂O ≥ 17.5.

[0017] According to another optional feature of the invention, the composition includes

%SiO ₂	62-68
%Al ₂ O ₃	1.5-2.5
%Na ₂ O	> 17
%K ₂ O	0-3
%CaO	5-8
%MgO	< 3
%B ₂ O ₃	5.5-6.5.

[0018] According to another optional feature of the invention, the composition includes

%SiO ₂	62-66.5
%Al ₂ O ₃	1.5-2.9
%Na ₂ O	≥ 17.5
%K ₂ O	0.1-1.0
%CaO	5.0-7.0
%MgO	≤ 3.5
%B ₂ O ₃	4.7-7.0.

[0019] According to another optional feature of the invention, in the composition

%Na₂O+%K₂O 18.5-19.5

%CaO+%MgO 8-10.

[0020] According to another optional feature of the invention, in the composition

5 %Na₂O+%K₂O 17.9-21.1
%CaO+%MgO 7.95-9.25.

[0021] According to another optional feature of the invention, the composition satisfies the following equation:

$$10 \quad \%Na_2O + \%K_2O + \%CaO + \%MgO + \%B_2O_3 + \%BaO - 2\%Al_2O_3 \leq 33.$$

[0022] According to another optional feature of the invention, the composition satisfies the following equation:

$$15 \quad \%Na_2O + \%K_2O + \%CaO + \%MgO + \%B_2O_3 + \%BaO - 2\%Al_2O_3 \leq 32.$$

[0023] According to another optional feature of the invention, the composition has a maximum liquidus temperature (viscosity = 10^{2.5} poise) comprised between 1.050 and 1.170 °C, a drop temperature (viscosity = 10³ poise) comprised between 950 and 1.070 °C and a devitrification temperature comprised between 650 and 850 °C. –

[0024] According to another optional feature of the invention, the composition has a maximum liquidus temperature (viscosity = 10^{2.5} poise) comprised between 1.050 and 1.150 °C, a drop temperature (viscosity = 10³ poise) comprised between 950 and 1.050 °C and a devitrification temperature comprised between 700 and 780 °C.

[0025] According to another optional feature of the invention, the composition is one of the following:

- 25 - SiO₂ 64.29 %; Na₂O 18.12 %; K₂O 0.47 %; CaO 6.07 %; MgO 2.97 %; B₂O₃ 5.95 %; and Al₂O₃ 1.85 % (C₆);
- SiO₂ 64.37 %; Na₂O 18.46 %; K₂O 0.34 %; CaO 5.68 %; MgO 2.95 %; B₂O₃ 6.01 %; and Al₂O₃ 1.95 % (C₇);
- SiO₂ 64.28 %; Na₂O 19.02 %; K₂O 0.41 %; CaO 5.34 %; MgO 2.72 %; B₂O₃ 5.91 %; and Al₂O₃ 2.11 % (C₈);
- 30 - SiO₂ 66.3 %; Na₂O 17.6 %; K₂O 0.52 %; CaO 5.95 %; MgO 2.82 %; B₂O₃ 4.8 %; and Al₂O₃ 1.75 % (C₉);
- SiO₂ 64.86 %; Na₂O 18.06 %; K₂O 0.51 %; CaO 5.94 %; MgO 2.94 %; B₂O₃ 5.52 %; and Al₂O₃ 2.02 % (C₁₀);
- 35 - SiO₂ 63.24 %; Na₂O 17.94%; K₂O 0.38 %; CaO 6.01 %; MgO 3.02 %; B₂O₃ 6.74 %; and Al₂O₃ 2.48 % (C₁₁);
- SiO₂ 63.05 % Na₂O 19.41 %; K₂O 0.44 %; CaO 6 %; MgO 2.96 %; B₂O₃ 5.49 %; and Al₂O₃ 2.51 % (C₁₂);
- SiO₂ 62.28 %; Na₂O 20.11 %; K₂O 0.41 %; CaO 5.97 %; MgO 2.76 %; B₂O₃ 5.41 %; and Al₂O₃ 2.8 % (C₁₃);

40 further including up to about 2 % in weight of impurities and other conventional compounds such as Fe₂O₃, but P₂O₅ and SO₃ being excluded from all of said compositions.

[0026] The scope of the invention also embraces the glass wool products that include fibres having the above mentioned compositions.

45 Brief description of the drawings

[0027] In the enclosed sheet of drawings:

Fig. 1 is a very schematic representation of a test apparatus used for calculating the dissolution rate constant, K_{SiO₂}, of the Compositions C₆ to C₁₃ according to the invention;

Fig. 2 is a graph where the dissolution rate constant K_{SiO₂} (in ng of SiO₂*cm⁻²*h⁻¹) of each one of the Compositions C₆ to C₁₃ according to the invention are represented, as a function of the analysis time, in hours; and

Fig. 3 is a graph where the behaviour of the dissolution rate constant K_{SiO₂} of each of the Compositions C₇ to C₁₃ according to the invention, with respect to the dissolution rate constant of Composition C₆, is represented, as a function of the analysis time, in hours.

[0028] The invention will be further disclosed by means of the Reference Compositions C₁ to C₅ and the Compositions C₆ to C₁₃, which represent Examples of the invention and whose formulations and test results are shown in Table

I.

Preliminary tests

- 5 [0029] Preliminary tests were carried out, with and without the use of P_2O_5 and SO_3 . The following Reference Compositions C_1 to C_5 were chosen because of their significance.

Reference Compositions C_1 to C_3

- 10 [0030] In a first series of tests, the Reference Compositions C_1 , C_2 and C_3 (see Table I) are prepared. In all of them P_2O_5 is included, and a relatively high content of R_2O ($\geq 17.65\%$) and a relatively low content of RO ($\leq 10.45\%$) are maintained, while variable and conventional amounts of B_2O_3 , SO_3 and ferric oxide (Fe_2O_3) are used. In Reference Composition C_1 , the Al_2O_3 content is about twice as much as in the Reference Compositions C_2 and C_3 .

- 15 [0031] It can be seen in Table I (row ($T_{1/2}$)) that by maintaining a relatively high SiO_2 content, in the Reference Compositions C_1 , C_2 and C_3 aforementioned:

1. Decrease of Al_2O_3 percentage enhances biosolubility.
2. Increase of P_2O_5 and SO_3 enhances biosolubility.
3. Maintaining of a high content of R_2O enhances biosolubility.

- 20 [0032] The mechanical strength of the fibres (especially, in Reference Compositions C_2 and C_3) was remarkably lower than the strength of fibres obtained with conventional non-biosoluble compositions.

Reference Compositions C_4 and C_5

- 25 [0033] Two other biosoluble Reference Compositions C_4 and C_5 are prepared. In Reference Composition C_4 a relatively high content of Al_2O_3 (2.32 %), a relatively low content of R_2O (15.69 %) and a relatively high content of RO (10.8 %) are used, and in both of them conventional amounts of Fe_2O_3 are included. In view of the relatively high Al_2O_3 content, the B_2O_3 content is increased proportionally to improve biosolubility without impairing workability. In Reference Composition C_5 a relatively low content of Al_2O_3 (1.72 %), a relatively high content of R_2O (17.53 %) and relatively low content of RO (10.12 %) are used. In neither of these Reference Compositions P_2O_5 or SO_3 is included. In Table I the test results can be seen, leading to the following conclusions:

4. Biosoluble glasses can be obtained without P_2O_5 or SO_3 .
- 35 5. The suitability of maintaining the sum of alkali oxides high, with the corresponding decrease of alkali-earth oxides, is confirmed.

- 40 [0034] The mechanical strength of the fibres of Reference Compositions C_4 and C_5 was much higher than the strength of fibres obtained with the Reference Compositions C_1 to C_3 .

Preferred embodiments of carrying out the invention

- 45 [0035] Going on from the above conclusions, in a new series of tests, Compositions C_6 to C_{13} according to the invention are prepared. In all of them, the R_2O content is increased (in detriment of RO), and the Al_2O_3 content is also increased. Of course, according to the above mentioned guidelines P_2O_5 and SO_3 are excluded.

Compositions C_6 to C_8 according to the invention

- 50 [0036] The change in chemical composition resides in a progressive increase in Al_2O_3 (1.85 %, 1.95 % and 2.11 %). On the other hand, this increase is tried to be balanced by a corresponding increase of Na_2O (18.12 %, 18.46 % and 19.02 %) in detriment of a progressive decrease of RO. The dissolution rate constant, K_{SiO_2} , clearly decreases from C_6 to C_8 . The amount of Na_2O added is not sufficient to counteract the effect of Al_2O_3 in the stabilisation of the crystal network. The decrease of RO fails to counteract this effect of the increase in Al_2O_3 .

Compositions C_9 to C_{11} according to the invention

- [0037] The strategy pursued has been to gradually increase the Al_2O_3 content (1.75 %, 2.02 % and 2.48 %), increasing at the same time the amount of B_2O_3 (4.80 %, 5.52 % and 6.74 %). The analysis of the dissolution rate con-

stant, K_{SiO_2} , shows a decreased value provoked by the increase in Al_2O_3 . The B_2O_3 added is not able to totally balance the increase of Al_2O_3 produced from C_9 to C_{11} .

[0038] However, the values of the dissolution rate constants, K_{SiO_2} , for C_{10} and C_{11} are higher than for C_8 which means that in an *in vivo* biosolubility test the result would be favourable.

5 [0039] The constant K_{SiO_2} obtained for C_9 is consistent with that obtained for C_6 . The constant of C_6 is slightly more favourable because C_6 contains a little more B_2O_3 .

Compositions C_{12} and C_{13} according to the invention

10 [0040] In these latter compositions exemplifying the invention the content of Al_2O_3 and of Na_2O is extremely high, while the B_2O_3 content is relatively low. The dissolution rate constants, K_{SiO_2} , are consistent with each other and determined by the Al_2O_3/Na_2O ratio. Composition C_{12} would be biosoluble in an *in vivo* test because its dissolution rate constant value is higher than that obtained in Composition C_8 .

15 Discussion of results in Table I:

[0041] In Table I the formulations and the test results of Reference Compositions C_1 to C_5 and Compositions C_6 to C_{13} according to the invention are shown. These results allow to deduce that:

- 20 1. The Al_2O_3 effect clearly impairs the biosolubility;
 2. Elements which most enhance biosolubility are Na_2O , which enhances the dissolution of the vitreous network, and B_2O_3 , which does likewise but in a more moderate extension;
 3. Although glasses of Compositions C_6 to C_{13} according to the invention have a KI value close to and even lower than 30, its estimated $T_{1/2}$ value and/or its biosolubility value are very favourable;
 25 4. Compositions C_6 to C_{13} of the invention provide temperature ranges for fibre production ($F_{fibr.}$) well adapted to current manufacture methods and apparatuses;
 5. Compositions C_6 to C_{13} of the invention present devitrification temperatures ($Temp_{dev.}$) more favourable than Reference Compositions.

30 [0042] Mechanical strength values of Compositions C_6 to C_{13} according to the invention were much higher than the respective values of Reference Compositions C_1 to C_5 .

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Table I

	Reference Compositions						Compositions according to the invention						
	C ₁	C ₂	C ₃	C ₄	C ₅	C ₆	C ₇	C ₈	C ₉	C ₁₀	C ₁₁	C ₁₂	C ₁₃
SiO ₂	63.98	64.6	65.8	65.1	64.6	64.29	64.37	64.28	66.3	64.86	63.24	63.05	62.28
Na ₂ O	17.8	17	17.3	14.4	16.6	18.12	18.46	19.02	17.6	18.06	17.94	19.41	20.11
K ₂ O	0.43	0.65	0.55	1.29	0.93	0.47	0.34	0.41	0.52	0.51	0.38	0.44	0.41
CaO	7.8	7.7	7.35	7.13	6.6	6.07	5.68	5.34	5.95	5.94	6.01	6	5.97
MgO	2.65	2.55	2.4	3.67	3.52	2.97	2.95	2.72	2.82	2.94	3.02	2.96	2.76
B ₂ O ₃	4.2	5.8	4.2	6	5.93	5.95	6.01	5.91	4.80	5.52	6.74	5.49	5.41
P ₂ O ₅	0.81	0.2	1	0	0	0	0	0	0	0	0	0	0
SO ₃	0.32	0.41	0.41	0	0	0	0	0	0	0	0	0	0
Fe ₂ O ₃	0.12	0.11	0.11	0.11	0.09	0.28	0.24	0.21	0.26	0.15	0.19	0.14	0.26
Al ₂ O ₃	1.95	1	0.9	2.32	1.72	1.85	1.95	2.11	1.75	2.02	2.48	2.51	2.8
R ₂ O	18.23	17.65	17.85	15.69	17.53	18.59	18.8	19.43	18.12	18.57	18.32	19.85	20.52
RO	10.45	10.25	9.75	10.80	10.12	9.04	8.63	8.06	8.77	8.88	9.03	8.96	8.73
R ₂ O/RO	1.74	1.72	1.83	1.45	1.73	2.06	2.18	2.41	2.07	2.09	2.03	2.22	2.35
Temp _{liq}	1115.0	1091.5	1134.0	1146.4	1114.0	1103.6	1097.9	1095.2	1140.37	1118.13	1077.81	1076	1066.6
Temp _{drop}	1019.4	1001.2	1037.5	1049.0	1016.4	1006.1	1000.1	996.6	1038.37	1018.89	983.74	978.02	958.81
Temp _{dev}	827.2	803.7	809.0	888.4	796.9	753.5	731.4	708.4	772.82	765.85	753.35	716.07	706.6
F _{flex}	192.20	197.50	228.50	160.60	219.50	252.6	268.7	288.2	265.55	253.04	230.39	261.95	262.21
KI	28.98	31.70	30.00	27.85	30.14	29.88	29.54	29.18	28.19	28.93	29.13	29.28	29.06
T _{1/2}	10.4	5.9	5	10.1	6.1	6.4	7	8.5	—	—	—	—	—
K _{SiO₂} (t=1 day)	—	—	—	—	—	159.4	147.3	114.7	162.65	119.3	117	136	111.8
K _{SiO₂} (t=2 days)	—	—	—	—	—	137.6	132	97.1	127.1	106.2	103.6	114.8	93.2
K _{SiO₂} (t=3 days)	—	—	—	—	—	135.3	123.4	87.1	116	97	94.8	106	80.4
K _{SiO₂} (t=4 days)	—	—	—	—	—	135.1	118.7	81.4	112.5	90.51	90.0	102.3	71.5

In Table I:

RO = Sum of oxides of elements pertaining to column 2A of the Periodic Table
R₂O = Sum of oxides of elements pertaining to column 1A of the Periodic Table

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Temp. _{liq.} =	Maximum liquidus temperature (viscosity = $10^{2.5}$ poise)
Temp. _{drop} =	Reference temperature for glass fiberisation or workability (viscosity = 10^3 poise)
Temp. _{dev.} =	Devitrification or crystal-growth temperature
F _{fibr.} =	Difference between Temp. _{drop} and Temp. _{dev.} . It is an indication of temperature ranges where fibre formation is possible with usual methods and apparatuses
T _{1/2} =	Half-life or time of elimination from the lung, in days, of 50 % of particles longer than 20 micron
K _{SiO2} =	SiO ₂ dissolution rate constant which will be discussed below in more detail; t = test days.

Calculation of glass dissolution rate constant K_{glass}

Conventional methodology:

[0043] The calculation of glass dissolution rate constant, K_{glass}, in different fluids is known; one of the most interesting tests in the mineral wool field is the one in which a glass composition is attacked with a fluid simulating, in terms of compounds and concentrations, lung fluid. This simulated biological fluid is also known as "Gamble's solution" and its composition is the following:

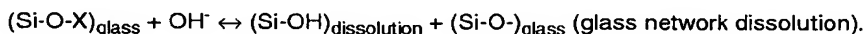
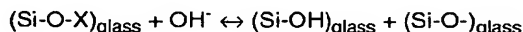
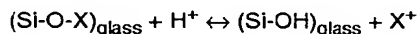
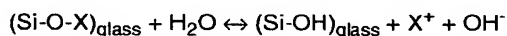
NaCl	6.415 g/l
NaHCO ₃	32.70 g/l
CaCl	20.19 g/l
Na ₂ HPO ₄ .12H ₂ O	0.358 g/l
Na ₂ SO ₄	0.079 g/l
MgCl ₂ .6H ₂ O	0.212 g/l
Glycine	0.118 g/l
Na _x Citrate+tartrate+lactate+pyruvate	0.680 g/l
Formaldehyde	1.000 ml/l

[0044] In the conventional methodology, some variations of this fluid are also used.

[0045] In prior art, this test has been carried out on glass fibres having a well controlled distribution of diameter and length, in order to calculate the specific surface in the most precise way possible. The test implies a separation and characterization step of the fibres. This is a long, tedious and expensive procedure and requires a sophisticated instrumentation.

[0046] The test is carried out controlling three instrumental parameters: pH, temperature and flow rate.

[0047] The balances of dissolution reactions in the test are the following:



[0048] These reactions are produced at a surface level and usually the dissolution constant K_{glass} is expressed as nanograms of glass dissolved per square centimetre and hour (ng glass*cm⁻²*h⁻¹).

Methodology used for the calculation of the dissolution rate constant K_{SiO2} of SiO₂ in the Compositions C₆ to C₁₃ according to the invention:

[0049] Using the aforementioned conventional methodology but modified as said below, tests have been carried out to estimate the SiO₂ dissolution rate constant, K_{SiO2}, in order to observe its evolution as glass composition is changed.

[0050] Starting from the fact that the half-lives in the lung or *in vivo* of Compositions C₆ to C₈ according to the invention are known (see values of row "T_{1/2}" in Table I):

- Composition C₆ showed to be biosoluble (T_{1/2} = 6.4) by means of an inhalation *in vivo* test, since the half-life was remarkably lower than 10 days (Directive 97/69/CE). The value of the dissolution rate constant, with t = 4 days, for this composition is 135.1 ng SiO₂*cm⁻²*h⁻¹. This value is used as the highest reference value.
- Composition C₈ showed to be the most unfavourable one (T_{1/2} = 8.5) as far as the half-life period, by inhalation, is

concerned. The value of the dissolution rate constant, with $t = 4$ days, for this Composition C₈ is $81.4 \text{ ng SiO}_2 \cdot \text{cm}^{-2} \cdot \text{h}^{-1}$ and is used as the lowest reference value.

[0051] Although a good correlation between *in vitro* dissolution constant and the period of half-life has not been proved, said constant can serve as an indication when the real chemical behaviour in the lungs of a living being is evaluated.

[0052] The conventional methodology to calculate the dissolution constant, K_{glass} , in Gamble's solution, has been modified by the inventors to achieve a quicker and simpler method allowing the comparison of different compositions, although not intended to establish values of dissolution constants directly comparable to those obtained through the conventional methodology used with fibres.

[0053] The main differences between both methodologies are the manner in which the sample is introduced into the test apparatus and the duration of the test.

[0054] In the methodology used here, a glass powder sample with a known extremely high (about $1 \text{ m}^2/\text{g}$) specific surface has been used in order to eliminate the problems that arise when --as happens in the conventional methodology-- the comparison between two compositions of fibres that have different diameters and lengths is needed. With the glass powder, it is possible to avoid the expensive step of the conventional methodology for separating and characterizing the fibres.

[0055] With the methodology used here, only the chemical attack of fluid on the glass powder is evaluated.

[0056] The tests are carried out in a relatively short period of time, from about 96 to 120 hours.

[0057] The test procedure (which does not form part of the invention) is described hereafter with reference to Fig. 1:

[0058] The fluid (Gamble's solution) 1, contained in a tank 2, is continuously corrected, in order to obtain a constant pH value, by means of CO₂ bubbling 3. A peristaltic pump 4 draws fluid to the sample cell 5, which contains the glass powder sample and which is maintained at a constant temperature (Temp.) with a thermostatic bath 6. The fluid portion with solved glass is collected in bottles at the outlet of the sample cell and analyzed by atomic absorption in 7.

[0059] The pH, temperature and flow rate values chosen for the tests of the glass compositions are the following:

pH: 7.6 ± 0.1

Temp.: $37 \pm 0.5 \text{ }^\circ\text{C}$

Flow rate: 140-180 ml/day.

[0060] The test results are presented, partly, in the above Table I and, partly, in Figs. 2 and 3, described above.

[0061] Although the invention has been described with reference to specific embodiments, the description is intended to be illustrative of the invention and is not intended to be limiting. A number of modifications and applications may occur to those skilled in the art without departing from the scope of the invention as defined in the appended claims. Particularly, it is understood that the manufacturers in this technical field experience problems in composition variation from one raw material batch to another one, which make admissible tolerances in $\pm 2 \%$ of absolute value, in some of the components of a given formulation. For this reason, in spite of the apparent precision with which the weight percentages of the invention compositions are given, the skilled in this art must understand that such values are nominal and that the claims also cover the compositions that respond to said nominal values affected by the usual tolerances in this technical field.

Claims

1. A biosoluble composition of glass fibres for the production of glass wools and the like, characterized in that it includes the following compounds in the specified weight percentages of the whole composition:

%SiO ₂	60-70
%Al ₂ O ₃	1-3
%Na ₂ O+%K ₂ O	> 17
%CaO	≤ 8
%MgO	2-4
%CaO+%MgO	< 10.5
%R ₂ O/%RO	> 1.65
%B ₂ O ₃	4.5-8,

where RO is the sum of oxides of elements pertaining to column 2A of the Periodic Table and R₂O is the sum of oxides of elements pertaining to column 1A of the Periodic Table, the composition further including up to about 2 % in weight of impurities and other conventional compounds such as Fe₂O₃, but P₂O₅ and SO₃ being excluded

from the composition.

2. The composition according to claim 1, characterized in that it includes 60-68 % of SiO_2 and in that $\% \text{Na}_2\text{O} + \% \text{K}_2\text{O} \geq 17.5$.

3. The composition according to claim 1, characterized in that it includes

$\% \text{SiO}_2$	62-68
$\% \text{Al}_2\text{O}_3$	1.5-2.5
$\% \text{Na}_2\text{O}$	> 17
$\% \text{K}_2\text{O}$	0-3
$\% \text{CaO}$	5-8
$\% \text{MgO}$	< 3
$\% \text{B}_2\text{O}_3$	5.5-6.5.

4. The composition according to any of claims 1 and 2, characterized in that it includes

$\% \text{SiO}_2$	62-66.5
$\% \text{Al}_2\text{O}_3$	1.5-2.9
$\% \text{Na}_2\text{O}$	≥ 17.5
$\% \text{K}_2\text{O}$	0.1-1.0
$\% \text{CaO}$	5.0-7.0
$\% \text{MgO}$	≤ 3.5
$\% \text{B}_2\text{O}_3$	4.7-7.0.

5. The composition according to any of claims 1 to 4, characterized in that

$$\% \text{Na}_2\text{O} + \% \text{K}_2\text{O} 18.5-19.5$$

$$\% \text{CaO} + \% \text{MgO} 8-10.$$

6. The composition according to any of claims 1 to 4, characterized in that

$$\% \text{Na}_2\text{O} + \% \text{K}_2\text{O} 17.9-21.1$$

$$\% \text{CaO} + \% \text{MgO} 7.95-9.25.$$

7. The composition according to any of claims 1 to 6, characterized in that it satisfies the following equation:

$$\% \text{Na}_2\text{O} + \% \text{K}_2\text{O} + \% \text{CaO} + \% \text{MgO} + \% \text{B}_2\text{O}_3 + \% \text{BaO} - 2 * \% \text{Al}_2\text{O}_3 \leq 33.$$

8. The composition according to claim 7, characterized in that it satisfies the following equation:

$$\% \text{Na}_2\text{O} + \% \text{K}_2\text{O} + \% \text{CaO} + \% \text{MgO} + \% \text{B}_2\text{O}_3 + \% \text{BaO} - 2 * \% \text{Al}_2\text{O}_3 \leq 32.$$

9. The composition according any of claims 1 to 8, characterized in that it has a maximum liquidus temperature (viscosity = $10^{2.5}$ poise) comprised between 1.050 and 1.170 °C, a drop temperature (viscosity = 10^3 poise) comprised between 950 and 1.070 °C and a devitrification temperature comprised between 650 and 850 °C.

10. The composition according to claim 9, characterized in that it has a maximum liquidus temperature (viscosity = $10^{2.5}$ poise) comprised between 1.050 and 1.150 °C, a drop temperature (viscosity = 10^3 poise) comprised between 950 and 1.050 °C and a devitrification temperature comprised between 700 and 780 °C.

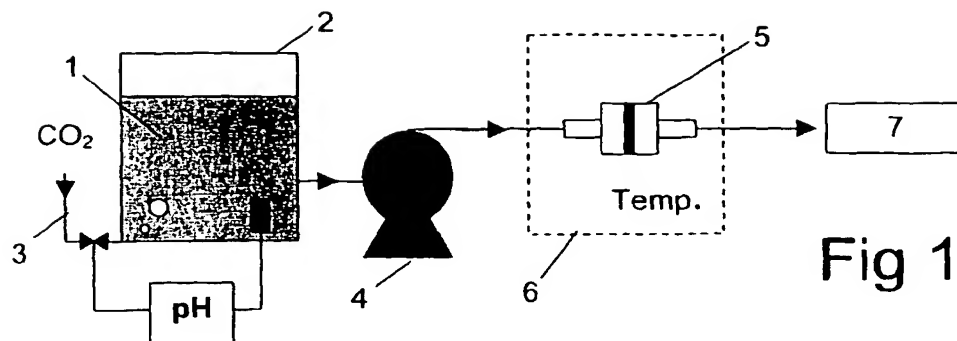
11. The composition according to one of claims 1 to 10, characterized in that it is one of the following:

- SiO_2 64.29 %; Na_2O 18.12 %; K_2O 0.47 %; CaO 6.07 %; MgO 2.97 %; B_2O_3 5.95 %; and Al_2O_3 1.85 % (C_6);
- SiO_2 64.37 %; Na_2O 18.46 %; K_2O 0.34 %; CaO 5.68 %; MgO 2.95 %; B_2O_3 6.01 %; and Al_2O_3 1.95 % (C_7);
- SiO_2 64.28 %; Na_2O 19.02 %; K_2O 0.41 %; CaO 5.34 %; MgO 2.72 %; B_2O_3 5.91 %; and Al_2O_3 2.11 % (C_8);

- SiO₂ 66.3 %; Na₂O 17.6 %; K₂O₃ 0.52 %; CaO 5.95 %; MgO 2.82 %; B₂O₃ 4.8 %; and Al₂O₃ 1.75 % (C₉);
- SiO₂ 64.86 %; Na₂O 18.06 %; K₂O 0.51 %; CaO 5.94 %; MgO 2.94 %; B₂O₃ 5.52 %; and Al₂O₃ 2.02 % (C₁₀);
- 5 - SiO₂ 63.24 %; Na₂O 17.94 %; K₂O 0.38 %; CaO 6.01 %; MgO 3.02 %; B₂O₃ 6.74 %; and Al₂O₃ 2.48 % (C₁₁);
- SiO₂ 63.05 %; Na₂O 19.41 %; K₂O 0.44 %; CaO 6 %; MgO 2.96 %; B₂O₃ 5.49 %; and Al₂O₃ 2.51 % (C₁₂);
- 10 - SiO₂ 62.2 8%; Na₂O 20.11 %; K₂O 0.41 %; CaO 5.97 %; MgO 2.76 %; B₂O₃ 5.41 %; and Al₂O₃ 2.8 % (C₁₃);

further including up to about 2 % in weight of impurities and other conventional compounds such as Fe₂O₃, but P₂O₅ and SO₃ being excluded from all said compositions.

12. A glass wool product, wherein fibres of the composition defined in claims 1-11 are included.



$\text{ng SiO}_2 \cdot \text{cm}^{-2} \cdot \text{h}^{-1}$

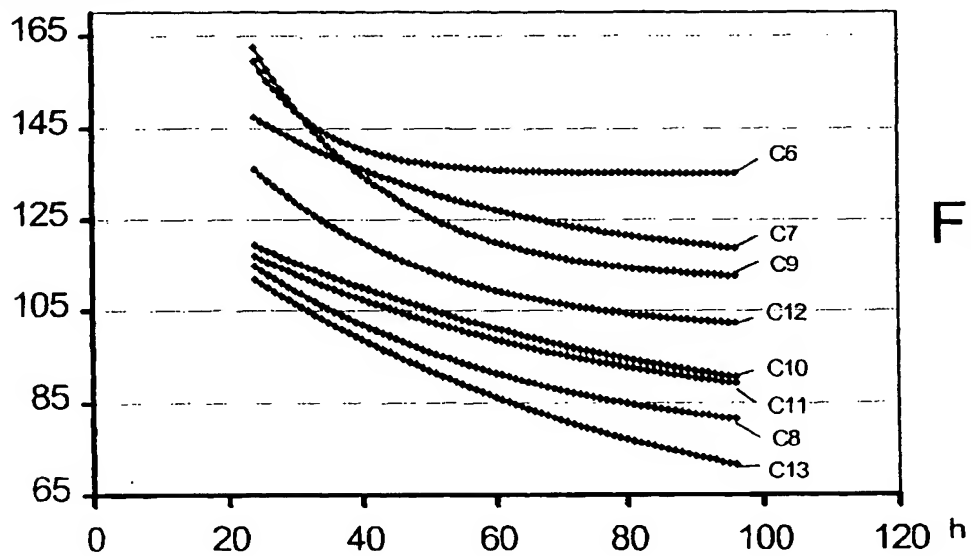


Fig 2

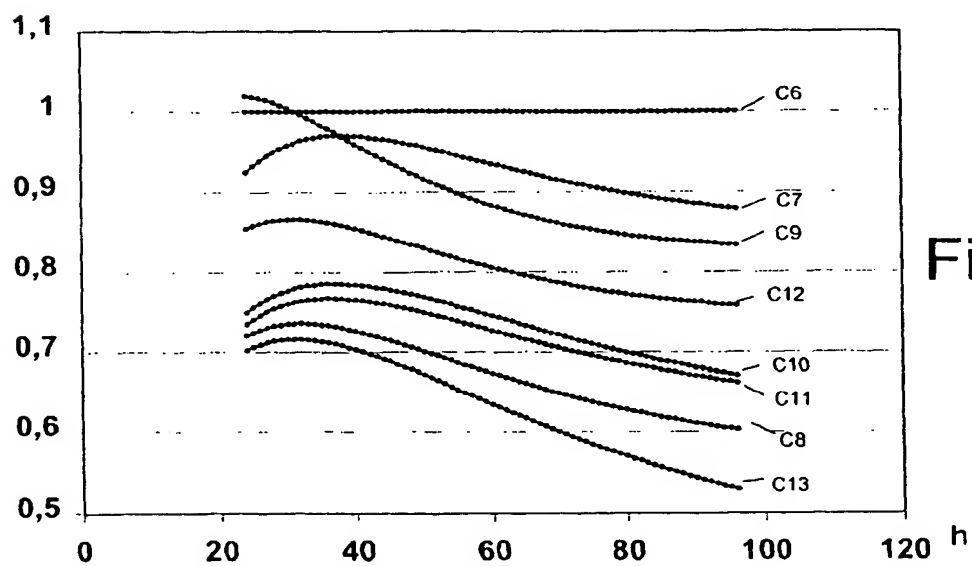


Fig 3



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